GENERAL SURVEY OF ATMOSPHERIC DIFFUSION.

A.S.MONIN

1. Significance of Atmospheric Diffusion as a Science.

The object of atmospheric diffusion as a science is the study of pollution propagation in the air. One of the most important practical problems which requires developing this branch of science is the air pollution by industry and transport, primarily urban pollution. If atmospheric diffusion did not exist in the air the pollution would accumulate in the lower layer of atmosphere and the inhabitants of the cities would not be able to breathe without gas-masks. Of no less importance is the problem of distribution of radioactive matter, which lately became a problem of great trouble to humanity. Due to atmospheric diffusion everybody of us, to a certain extent, is subjected to the effect of radioactivity - a result of atomic explosions.

We come across the phenomenon of atmospheric diffusion in agriculture when the plants are chemically pollinated in the struggle against pests or when they are defended from frosts by producing a smoke. The sea salt and the volcanic dust, bacteria and viruses, pollen and seeds of plants are distributed in the air due to atmospheric diffusion. The air masses are saturated with humidity above the sea and with dust above the deserts due to the same cause.

The study of atmospheric diffusion is of great importance for practical purposes as well as for adjacent branches of science. And at the same in view of the intriguing complexity of the studied phenomena the investigation of them can give an aesthetical satisfaction even to the most exacting scientists. It mostly refers to the specialists in hydrodynamics and geophysics who are engaged in the problems related to atmospheric diffusion.

2. Factors Influencing on Atmospheric Diffusion.

Atmospheric diffusion is a very complex phenomenon which is dependent on many factors. First, one needs know the way the pollutions come into the air, in other words, the nature of a source. The pollution entering the air may be produced by industrial enterprises, artificial sources and the earth's surface itself. The source may be instantaneous and continuous with a constant or variable productivity. The source may be a point source (ground or elevated) or distributed along the line, over the surface or volume. It is also important to know whether the the pollution particles have a certain speed at the outlet of a source (for instance, the exhaust speed of a gas at the outlet of a chimney) and what temperature of the polluted air is at theoutlet of a source (the heated gas as compared to the surrounding air will flow up, the cooled one- down).

Secondly, one needs know the peculiarities of the pollution propagating throughout the air under different meteorological conditions. The air pollutions are transported by the air flows and they diffuse also due to turbulence.The object of hydrodynamics is to describe these processes.

To describe the transfer of the pollution by wind one has to know kinematics of the air flows. In particular, to estmate the propagation of the pollution in the surface layer it is necessary to have the information about the vertical wind profile under different meteorological conditions (particularly - under different thermal stratification of the air). To estimate an average pollution around a given source for a long period of time it is necessary to have statistical data about a wind direction and a wind speed in the given region. To estimate the pollution from the instantaneous point on the global scale one needs know kinematics of the air flows of synoptic scale over a great part of the globe and for a sufficiently long period of time (measured in weeks). Besides regular macroscopic flows there exist chaotic hydrodynamic motions of different scales up to a very small scale of the order of 1 cm. Those chaotic motions are called turbulence. Mixing process due to the turbulence is the reason for turbulent diffusion of pollutions. In order to describe the turbulent diffusion one needs know some statistical characteristics of the turbulent velocity field. These characteristics, generally speaking, appear to be dependent upon meteorological conditions and mainly upon the field of averaged wind velocity and thermal stratification of the air. For example, under stable stratification the turbulent diffusion is going on very slowly and the pollutions are transported by wind almost without dispersion. On the contrary, under convection the turbulent diffusion leads to a rapid dispersion of pollutions.

The third group of factors influencing on the atmospheric diffusion lies in the properties of pollution itself. Primarily, it is necessary to know what effect the gravity produces on the pollution. Gases which are heavier than the air and comparatively large particles fall down. The speed of the fall of particles depends on their size, specific weight and form. The possibility of chemical and radioactive transformation of pollution as well as physical transformation such as coagulation, sublimation and absorption on aerosoles also should be taken into account. In particular, the interaction of the pollution with atmospheric humidity as water vapour, water drops in clouds and fogs and precipitation may be essential. For instance, the rains may clear the air from pollutions making them to fall to the earth's surface.

The fourth group of factors lies in the pollution interaction with the earth's surface or water surface. The pollution may be absorbed by the surface (water surface can absorb the majority of pollutions). The pollution may also be reflected from the surface and come back into the air. The intermediate cases are also possible. There are cases of partial absorption and reflection or absorption for a certain (random) period of time after which the pollution raises up into the air. When the boundary conditions for the pollution on the earth's surface are formulated mathematically it is necessary to take into account its roughness and its ability to absorb the pollution of a given kind. Some obvious complications will arise as a result of the inhomogeneities of the earth's surface such as varieties of a relief, presence of houses and trees. The investigations in the theory of atmospheric diffusion are directed to the development of standard methods of estimation of the air pollution under idealized average conditions (usually over a flat relief under stationary atmospheric conditions) as well as to the study of influence on atmospheric diffusion of one or another enumerated above factors (for instance, the influence of thermal stratification of air). I shall mostly touch on the investigations of the theory of turbulent diffusion.

I don't mean to review the literature on this question and confine myself to mentioning only few authors. I am greatly pleased to speak here in England about the outstanding results of the English scientists such as Richardson, Taylor, Sutton, Batchelor and many others.

3. A Specific Feature of Turbulent Diffusion.

A specific feature of turbulent diffusion is a wide spectrum of scales of turbulent motions giving rise to the air mixing process. The character of turbulent diffusion depends upon the distribution of the energy among turbulent motions of different scales. The greatest of them can be called the scale of turbulence ℓ . The velocities at points the distance between which does not exceed ℓ are statistically connected. Therefore the pollution particles the distance between which does not exceed ℓ , will move, generally speaking, independently of each other. This breaks the analogy between turbulent and molecular diffusion.

In a number of cases the scale of turbulence ℓ appears to be small as compared to the size of the region in which the diffusion actually occurs (for instance, as compared to the diameter $\mathcal{L}$ of the pollution cloud). In these cases one can speak of the diffusion in the field of small scale turbulence. In such cases the pollution particles at comparatively small (as compared to $\mathcal{L}$) distances move independently. The description of turbulent diffusion by analogy with that of molecular diffusion seems to be justifiable. Such an approach is usually applied to the description of turbulent diffusion along the vertical in the surface layer of atmosphere.

2. Analogy between the diffusion in the field of small scale turbulence and molecular diffusion.

The analogy lies in the following. The chaotic molecular motion can be characterized with an average velocity of molecular motion v_m (depending upon the gas temperature) and a molecule free path length ℓ_m. The coefficient of the molecular diffusion can be determined by means of the values $K_m \sim v_m \ell_m$. This coefficient is introduced into the theory as the coefficient of proportionality between the diffusion flux of a given substance S and the gradient of its concentration ∇s: $S = -\rho K_m \nabla s$ (ρ is the air density). Similarly, the chaotic turbulent motion can be characterized by the magnitude of turbulent fluctuation of the velocity v (which serves a measure of the intensity of turbulence) and the scale of turbulence ℓ (L.Prandtl in 1934 introduced " a mixing length" as ℓ, the value which is similar to a molecule free path length). Then the coefficient of turbulent diffusion $K \sim v \ell$ can be introduced. It is understood as the coeffidient of proportionality between an average turbulent flux of a given substance $S = \rho \overline{s' u'}$ and the gradient of its averaged concentration $\nabla \bar{s}$;

$$S = -\rho K \nabla \bar{s} \qquad (1)$$

(a bar denotes averaging and a dash denotes the deviation from the average value, u is the velocity field). Assumption of proportionality between S and $\nabla \bar{s}$ was formulated by W.Schmidt (1925). On the base of this assumption the so-called semi-empirical theory of turbulent diffusion was developed. In mathematical respect it is analogous to the theory of molecular diffusion in non-homogeneous medium.

The scales of molecular motions differ many times from those of turbulent motions. So, for molecular motions $v_m \sim 10^4$ cm/sec, $l_m \sim 10^{-5}$cm, $K_m \sim 10^{-1}$cm^2/sec in the surface layer of atmosphere, and for turbulent motions $v \sim 10$ cm/sec, $l \sim 10^2 - 10^3$cm, $K \sim 10^3 - 10^4$cm^2/sec. The figures show that the molecular processes can be neglected in the majority of problems of atmospheric diffusion. At the same time the difference in scales does not lead to the qualitative difference between the turbulent and molecular diffusion. More essential is the difference in the velocities of motions as the fitness of parabolic diffusion equation is more limited the less are the actual velocities of the motion of diffusing particles (this question will be considered below). It is also essential that in contrast to the mollecular diffusion the turbulent mixing in the atmosphere, as a rule, is non-isotropic. However, the corresponding generalization of theory does not present any difficulties (i.e. under l and K tensors should be understood).

5. A Semi-empirical Equation of Turbulent Diffusion.

A semi-empirical equation of turbulent diffusion for the surface layer of the atmosphere can be written as follows:

$$\frac{\partial S}{\partial t} + u\frac{\partial S}{\partial x} = \frac{\partial}{\partial x}K_x\frac{\partial S}{\partial x} + \frac{\partial}{\partial y}K_y\frac{\partial S}{\partial y} + \frac{\partial}{\partial z}K_z\frac{\partial S}{\partial z} \qquad (2)$$

Here the axis x is directed in the wind direction, the axis z is directed vertically, t is the time, u is the wind velocity, K_x, K_y, K_z are the coefficients of turbulent diffusion in the directions of x, y, z. If one needs take into account the gravitational fall of diffusing particles (with a velocity w) and the possible exponential decrease of the quantity of diffusing matter (with characteristic time $\ln 2/\alpha$) then the item $+ w\frac{\partial S}{\partial z} + \alpha S$ should be added to the left part of the equation (2). However, in the standard methods of calculation these items are usually not considered. The equation (2) is formulated for the half-space $z > z_0$, where z_0 is a roughness parameter of the earth's surface. For the surface $z = z_0$ one or another boundary condition is given for the concentration S. A typical problem of the equation (2) is to seek the solutions corresponding to instantaneous and continuous sources of pollution (in the investigation of continuous sources the item $\frac{\partial}{\partial x} K_x \frac{\partial S}{\partial x}$ as compared to $u \frac{\partial S}{\partial x}$ is usually neglected). The coefficients u, K_x, K_y, K_z of the ~~the~~ equation (2) are, generally speaking, variable. The analytical solution of the equation fitted for standard calculations one does manages to obtain only by some particular assumptions about these coefficients. So in the case of constant coefficients solutions of the equations (2) corresponding to the basic types of sources have been studied by O.F.T.Roberts as far back as in 1923. These solutions give a good qualitative description of diffusion processes. They don't agree however, quantitatively with experimental data (the rate of the concentration decreasing of the pollution as it removes from the source proves to be too small). Besides this the theory of the turbulent state in the surface layer of atmosphere as well as direct measurements of the coefficients of turbulent diffusion show that these coefficients are not constant. They increase with the height (under indifferent stratification they increase proportionally to the height). The solutions of the equation(2) when u = const and $K_y, K_z \propto z$ were studied by Bosanquet C.H. and Pearson J.I. in 1936. And, finally some authors considered the case when the wind velocity u and the coefficients of turbulent diffusion are proportional to some powers of the height z. The equations of this kind allow good approximation of the experimental laws. Sinse 1944 the similar methods were developed by D.L.Laikhtman(USSR) in detail in his works.

6. Statistical Approach towards Turbulent Diffusion.

The diffusion equation (2) can be deduced from the assumption that each individual diffusing particle moves randomly and its coordinates vary in time in accordance with the Markov random process. The equation (2) is the Focker-Plank equation for this random process. Such a deduction leads to a following statistical interpretation of the coefficients of turbulent diffusion

$$K_x = \frac{1}{2}\frac{d\sigma_x^2(t)}{dt}\,; \quad \sigma_x^2(t) = \overline{[x(t) - x(0)]^2} \qquad (3)$$

where $x(t)$ is the abscissa of a diffusing particle at the moment t (analogous equations are valid for K_y and K_z). Hence it appears that the primary concept is the dispersion of the coordinate of a diffusing particle (depending upon time) and not the coefficient of turbulent diffusion. The convenience of watching the moving particles (in other words the Lagrangian method of describing the medium motions and not the Eulerian one) is a specific feature of the theory of turbulent diffusion in contrast to the theories of other phenomena caused by turbulence. From this point of view the Lagrangian correlation function of the field velocity is the most convenient characteristic of turbulence

$$\overline{u_x(t)\,u_x(t+\tau)} = \overline{u_x^2}\;R_x(\tau) \qquad (4)$$

where $u_x(t) = \frac{dx(t)}{dt}$; is the x component of velocity of a diffusing particle at the moment t ; a bar denotes averaging in time.

An essential distinction of the statistical theory of turbulent diffusion from that of molecular diffusion lies in the supposition of existance of an instantaneous velocity of a particle.

Note, that there is an "evolution of a level" of a velocity field in turbulent medium; the time mean values of characteristics of a velocity field essentially depend upon the length of the interval averaging. That is why the detrmination of the correlation function (4) is valid, strictly speaking, only in the cases of small scale turbulence (in the sense indicated above). In a more general cases it is preferable to proceed from the Langrangian correlation function for the acceleration and not for the velocity.

The dispersion $\sigma_x^2(t)$ can be expressed by means of the correlation function (4) as follows

$$\sigma_x^2(t) = 2\overline{u_x^2}\int_0^t (t-\tau)R_x(\tau)\,d\tau \qquad (5)$$

This very important equation was proposed by G.I.Taylor (1921). O.G.Sutoon (1932) suggested that the function $R(\tau)$ should be approximated by the equation $R(\tau) = (1+\tau/T)^{-n}$. When t are large the equation $\sigma^2(t) \approx \frac{c^2}{2}(ut)^{2-n}$ is used; On this base O.G.Sutton obtained the equations for the concentration of the pollution corresponding to the basic types of sources. O.G.Sutton's equations proved to be very convenient for description of experimental data and became widely used for estimation of air pollution.

It follows from the Taylor's equation (5) that for small diffusion time $\sigma^2 \propto t^2$ and $K \propto t$; for large diffusion time $\sigma^2 \propto t$ and $K \rightarrow$ const (the last regularity is analogous to the case of molecular diffusion). Using this information and describing the concentration of the pollution in the presence of an instantaneous point with the help of a Gauss function having dispersions $\sigma_x^2(t), \sigma_y^2(t), \sigma_z^2(t)$ F.N.Frenkiel developed the methods for calculation of a diffusion of pollutions successfully used for description of urban pollution.

7. Influence of Thermal Stratification of Atmosphere on Turbulent Diffusion.

As it was already mentioned, the turbulent diffusion in the lower layers of atmosphere is essentially dependent on thermal stratification of air. It is convenient to use a dimensionless parameter while the influence of stratification on the turbulent regime is taken into account

$$Ri = \frac{g}{\theta} \frac{\partial \theta / \partial z}{(\partial u / \partial z)^2} \tag{6}$$

where g is the acceleration of gravity, and θ is the so-called potential temperature (in the lower layer of atmosphere $\theta \approx T + \Gamma z$, where Γ is the usual temperature and $\Gamma \approx 1^\circ C / 100\,m$). This parameter was introduced by L.F.Richardson (1925). Ri is negative under thermal instability and positive under stable stratification. With the help of energetic considerations Richardson found out that if $Ri > Ri_{cr} > 0$ the turbulence decays losing its energy at the work against the Arkhimed forces. Lately, C.H.B.Priestly and E.L.Deacon in Australia, H.Lettau in the USA and A.M.Obukhov and A.S.Monin in the USSR developed similar methods for consideration of the influence of stratification on the turbulent state in the surface layer of atmosphere. Obukhov and Monin developed the similarity theory according to which the turbulent state is fully determined by three parameters - turbulent stress, vertical turbulent heat flux and a parameter g/θ, characterizing the influence of the Arkhimed forces. The influence of stratification upon the characteristics of a turbulent state are described with dimensionless multipliers depending on Ri which are efficiently determined in a number of cases.

9. Consideration of the Bounded Velocity of Turbulent Diffusion.

The considered theory of small scale turbulent diffusion has a disadvantage. It does not take into account that the velocity of pollution propagation in the turbulent atmosphere is bounded as the fluctuations of the wind velocity causing the turbulent mixing are bounded. The parabolic character of a semi-empirical diffusion equation means that the pollution leaving the source immediately propagates throughout the space and can be at once noticed at any large distance from the source. Usually this disadvantage tolerated as the volume inside of which the concentration of the pollution is not too small is always bounded and the distribution of pollution inside the volume is described with a parabolic diffusion equation, as a rule, satisfactorily. However, in some cases, (in particular, close to the actual boundaries of the pollution cloud) the use of a parabolic equation may lead to essential error. For example, the smoke going out of the chimney having the height h reaches the earth's surface at a distance of $\frac{u}{v}h$ from the chimney, where u is the wind velocity, v is the maximum velocity of smoke propagation along the vertical. At the same time in accordance with the solution of parabolic diffusion equation the smoke can be found at the earth's surface at any distance from the chimney.

The Soviet scientist Sheleikhovsky proposed the methods for calculation of propagation of the smoke out of the chimneys which are free of the disadvantage indicated above. These methods are on the use of a theory of a free turbulent jet. According to the Sheleikhovsky's equation the smoke going out of the chimney fills the cone. The axis of the cone lies in the direction of the wind, an angle of it depends upon the intensity of turbulence. Sheleikhovsky's equation makes it possible to determine only the average concentration in different cross sections of a smoke plume.

An efficient method is to generalize the diffusion equation in a way that it might become hyperbolic. Such a generalization was proposed by the Soviet scientist V.A.Fock in 1926, by E.S.Lyapin in 1948 and by S.Goldstein in 1951. In order to deduce one-dimensional hyperbolic diffusion equation one can proceed from the following assumptions: a) every individual diffusing particle moves randomly, b) and instantaneous velocity of a particle exists almost everywhere and is bounded, c) a particle coordinate and a direction of its motion form together a Markov random process.

The diffusion equation is odtained in the form

$$\frac{\partial s}{\partial t}+\frac{\partial S}{\partial z}=0; \quad \frac{1}{2a}\frac{\partial S}{\partial t}+S=-\frac{v^2}{2a}\frac{\partial s}{\partial z} \qquad (7)$$

where s is the concentration of particles, S is the turbulent flux of particles, v is the maximum velocity of particles, a is the characteristic frequency of turbulent fluctuations. Eliminating the turbulent flux one can obtain for the concentration of particles s from (7) the so-called telegraph equation. In the limit when $a \to \infty$, $v \to \infty$, $\frac{v^2}{2a} \to K$, a usual parabolic equation can be obtained.

10. Diffusion in the Field of Large Scale Turbulence.

When the scale of turbulence ℓ is not small as compared to the size of the pollution cloud, the regularities of turbulent diffusion are essentially different from those of molecular diffusion. For example, in contrast to molecular diffusion the velocity of changing the distance L between the two diffusing particles depends upon the distance L itself: the velocity, on average, is not large till L remains small and it grows large when L becomes large. It can be explained by the fact that the distance L essentially varies due to turbulent motions the scales of which are comparable with L. The small scale motions only slightly change this distance and the large scale ones carrying away both particles simultaneously transfer them without essential change of a distance between them. So the increase of the cloud size leads to the increase of an "effective diffusion coefficient".

Richardson was the first to pay attention to this phenomena in 1926. He suggested to describe this phenomena by a "distance neighbour function" $g(L,t)$ which is the probability density for distance L between two diffusing particles. Richardson suggested later that the change of the function $g(L,t)$ should be described by a parabolic diffusion equation with a diffusion coefficient K, depending on L. With the help of empirical data Richardson found out that $K(L) \propto L^{4/3}$. This law is valid for the phenomena of different scales beginning with the diffusion in the surface layer of atmosphere up to horizontal mixing in the scales of a general circulation of atmosphere.

Sutton's equations in which the diffusion coefficient increases with time qualitatively take into account Richardson's effect. However, the diffusion along the vertical in the surface layer of atmosphere takes place mostly due to small scale turbulence. At the same time turbulent motions of a very wide range of scales take part in horizontal mixing of air. For example, experiments of a continuous registration of a wind direction and observations of smoke plumes show that turbulent motions of large scales (some hundred meters and kilometers) leading to fluctuations of a wind direction and observations of smoke plumes show that turbulent motions of a large scale (some hundred meters and kilometers) leading to fluctuation of a wind direction with periods of several minutes have essential influence upon the diffusion of pollutions. Therefore the Richardson's effect should be necessarily taken into account ~~when describing~~ when describing horizontal mixing.

The Richardson's law $K(L) \propto L^{4/3}$ was explained by A.M.Obukhov (1941) as a consequence of similarity hypotheses of A.N.Kolmogoroff (1951) for turbulence with a large Reynolds numbers. According to the Kolmogoroff's hypothesis there exists the so-called inertial range of scales of turbulent motions. The statistical regime of these motions is fully determined by the effect of inertial forces which lead to transfer of energy of motions of large scales to those of smaller scales with a constant rate ε . The value ε which is equal to the dissipation of turbulent energy is the only parameter determining the turbulent regime within the inertial range. If the diffusion occurs as a result of turbulent motions of scales within the inertial range then $K(L) = C\varepsilon^{1/3} L^{4/3}$ (where C is a number) in other words we obtain the Richardson's law. The application of the similarity theory to turbulent diffusion was considered in detail in teresting works of Batchelor in 1950.

It is important to be able to estmate parctically the concentration of the pollution S . The knowlege of the "distance neighbour function" $g(L,t)$ for this purpose is not sufficien as S can be determined according g only in the case when the motions of diffusing particles are independent of each other. Some information of the concentration S in the presence of some or other sources can be given by the similarity theory. So, in the two dimensional case the distribution of concentration with regard to the centre of the pollution cloud in the presence of an instantaneous point source is as follows:

$$S(r,t) = \frac{Q}{\pi\varepsilon t^3} f\left(\frac{r^2}{\varepsilon t^3}\right) \qquad (8)$$

(r is the distance frpm the cloud centre). In particular, the cloud diameter grows proportionally to $t^{3/2}$.

In order to find a general form of the equation for the Richardson's diffusion, except the similarity theory, one can use the fact that the turbulence is homogeneous and isotropic in the registration system connected with the average motion of a medium. In this registration system under a given initial concentration $S_0(r)$ the concentration at the moment t can be determined as follows:

$$\tilde{S}(p,t) = a(\varepsilon^{1/3} p^{2/3} t)\, \tilde{S}_0(p) \qquad (9)$$

where the wave sign means Fourier transformation according to $\vec{x}$, ρ is the wave vector (p its magnitude), $a(\theta)$ is some dimensionless function which is equal to 1 if $\theta = 0$ and continuously decreasing to zero if $\theta \to \infty$. The choice of a concrete function $a(\theta)$ leads to a concrete diffusion equation. If one assumes that the distribution of concentration chages in time according to a semi-group law then $a(\theta)$ should be put equal to $e^{-c\theta}$.

Another approach to description of Richardson diffusion is proposed by A.M.Obukhov. It is suggested that the diffusion equation should be written in a six-dimensional space of coordinate and velocities by analogy with the theory of Brownian motion with inertia and with additional requirements of invariancy proceeding from the fact that the turbulence is locally homogeneous and isotropic.

11. Perspectives of further development of theory.

The possibilities of statistical theory of turbulent diffusion are far not exhausted. In the nearest future this theory, undoubtedly, will strongly progress following the progress of statistic theory of turbulence. Even now certain methods suggested for description of turbulence are to be used in the theory of turbulent diffusion. As an example I can indicate the present workes of Roberts in which the equations for correlation moments of the polltuion concentration and a velocity field are constructed with the help of the hydrodynamical equations. For the purpose of closing these equations the hypothesis of M.D.Millionshchikov is used. (i.e. the fourth moments are expressed by means of the second ones according to the equations which are valid for multidimensional norm l distribution). The method of characteristic functionals suggested by E.Hopf is another example.

In the present review I touched on the most general problems of atmospheric diffusion. I had no possibility to dwell on many interesting, particular questions to be discussed at our Symposium. I am confident of the successful work of the Symposium and hope it will serve a further progress of our science - the science of turbulent diffusion.